MOISTURE CURABLE COMPOSITIONS WITH ENHANCED ADHESION TO POLYOLEFINS

FIELD OF THE INVENTION:

[0001] The present invention relates to moisture curable compositions with enhanced adhesion to polyolefin substrates. More particularly, the present invention relates to moisture curable silicone polymer-containing compositions exhibiting enhanced adhesion to polyethylene and polypropylene substrates.

BRIEF DESCRIPTION OF RELATED TECHNOLOGY:

[0002] Alkoxysilylated polymers, in the presence of a catalyst, can be crosslinked by atmospheric moisture under ambient conditions. Compositions based on these types of polymers are often referred to as RTV adhesives (or sealants). The most well known example is the RTV silicone adhesives or sealants. Such silicone adhesives or sealants are generally durable and have excellent resistance to weathering and temperature extremes.

[0003] While cyanoacrylates, hot melts, epoxies, urethanes and two-part acrylics are commonly used as adhesives on plastic substrates, often times they are used with primers, in effect rendering them two-part adhesive systems. Physical treatments, such as surface roughening, plasma treatment, thermal treatment, among others, are other ways often used to improve adhesive wetting on polyethylene (PE) and polypropylene (PP) surfaces, but such treatments are often expensive and time consuming and do not necessarily provide the desired effect of enhanced bonding.

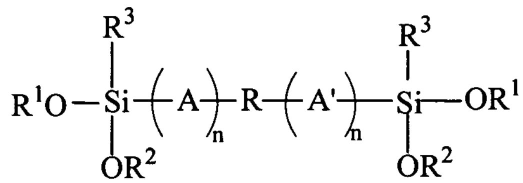
[0004] Alkoxysilylated polymers have not been generally used on plastic substrates, particularly polyethylene and polypropylene substrates, due to poor adhesive wetting characteristics on these substrates.

[0005] Thus, there is need for alkoxysilylated polymer-containing compositions with enhanced adhesion properties to plastic substrates, which do not require priming or physical surface treatment.

SUMMARY OF THE INVENTION:

[0006] Moisture curable compositions useful for adhesively bonding polyolefins, including polyethylenes and polypropylenes, include:

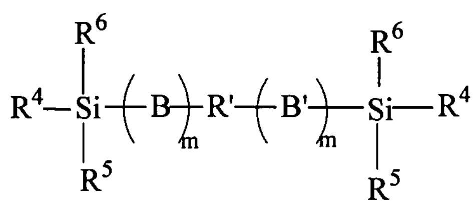
(a) alkoxysilyl capped polymer compounds within the following formula:



I

wherein R is a hydrocarbon diradical which may include heteroatom and/or silicone-containing groups or linkages; A and A' are each C_{1-30} linear or branched, substituted or unsubstituted aliphatic groups or aromatic-containing groups, with or without interruption by a carboxy, carbamate, carbonate, ureido, urethane or sulfonate linkage; n may be 0 or 1; R^1 and R^2 are substituted or unsubstituted C_{1-12} alkyl or aryl groups; R^3 is a C_{1-12} alkyl, alkenyl, alkoxy, aminoalkyl or aryl group, or a (meth)acryloxyalkyl group;

(b) at least one alkylsilyl capped plasticizer within the following formula:



II

wherein R' is a hydrocarbon diradical which may include heteroatom and/or silicone-containing groups or linkages; B and B' may be each C_{1-30} linear or branched, substituted or unsubstituted aliphatic groups or aromatic-containing groups, with or without interruption by a carboxy, carbamate, carbonate, ureido, urethane or sulfonate linkage; m may be 0 or 1; R^4 and R^5 are substituted or unsubstituted C_{1-12} alkyl or aryl groups; R^6 is a C_{1-12} alkyl, alkenyl, alkoxy,

aminoalkyl or aryl group, or a (meth)acryloxyalkyl group;

(c) an adhesion promoter;

(d) a filler present; and

(e) a moisture curing catalyst.

[0007] Polyether backbones, i.e., the "R" in Formula I above, including polypropylene oxide backbones, are useful for the alkoxysilyl capped polymer compounds of the present invention. Trialkoxysilyl capped polymer compounds, where "R^3" in Formula I above is a C_{1-12} alkoxy, are also useful.

[0008] Polyether backbones, i.e., the "R^1" in Formula II above, including polytetramethylene oxide backbones, are useful for the alkylsilyl capped polymer compounds of the present invention. Trialkylsilyl capped polymer compounds, where "R^6" in the Formula II above is a C_{1-12} alkyl, are also useful.

[0009] Desirably, the filler is a calcium carbonate filler present in from about 10 weight percent to about 70 weight percent on a total composition basis.

[0010] In another aspect of the present invention moisture curable compositions are provided that include:

(a) from about 5 weight percent to about 99 weight percent on a total composition basis of trialkoxysilyl capped polymer compounds within the following structure:

$$R^1O-\overset{\displaystyle R^3}{\underset{\displaystyle OR^2}{\overset{|}{\underset{|}{Si}}}}-\left(A\right)_n-R-\left(A'\right)_n-\overset{\displaystyle R^3}{\underset{\displaystyle OR^2}{\overset{|}{\underset{|}{Si}}}}-OR^1$$

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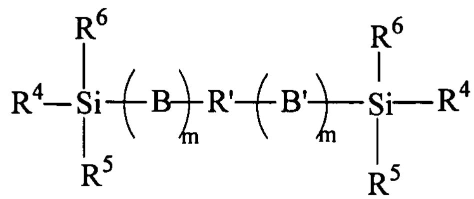
wherein R is a hydrocarbon diradical which may include heteroatom and/or silicone-containing groups or linkages; A and A' are each C_{1-30} linear or branched, substituted or unsubstituted aliphatic groups or aromatic-containing groups, with or without interruption by a carboxy, carbamate, carbonate, ureido, urethane or sulfonate linkage; n may be 0 or 1; R^1 and R^2 are

substituted or unsubstituted C_{1-12} alkyl or aryl groups; R^3 is a C_{1-12} alkoxy group;

(b) from about 0 weight percent to about 35 weight percent on a total composition basis of trialkylsilyl capped polymeric plasticizers within the following structure:



II

wherein R' is a hydrocarbon diradical which may include heteroatom and/or silicone-containing groups or linkages; B and B' may be each C_{1-30} linear or branched, substituted or unsubstituted aliphatic groups or aromatic-containing groups, with or without interruption by a carboxy, carbamate, carbonate, ureido, urethane or sulfonate linkage; m may be 0 or 1; R^4 and R^5 are substituted or unsubstituted C_{1-12} alkyl or aryl groups; R^6 is a C_{1-12} alkyl, alkenyl or aryl group;

(c) from about 1 weight percent to about 5 weight percent on a total composition basis of an aminopropyltrimethoxysilane adhesion promoter;

(d) from about 10 weight percent to about 70 weight percent on a total composition basis of a calcium carbonate filler; and

(f) a moisture curing catalyst.

[0011] Methods for bonding polyolefin substrates and articles so formed are also described.

BRIEF DESCRIPTION OF THE DRAWINGS:

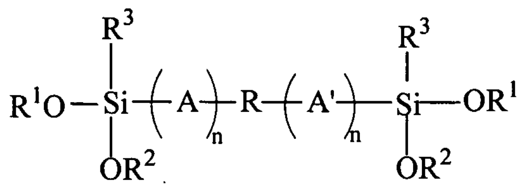
[0012] Figure 1 is a perspective view of a laminate having polyolefin substrates according to the present invention.

[0013] Figure 2 is a cross sectional view of the laminate of FIG. 1 showing a layer of inventive adhesive securely joining the polyolefin substrates.

DETAILED DESCRIPTION OF THE INVENTION:

[0014] Moisture curable compositions of the present invention exhibit enhanced adhesion to polyolefins, including polyethylene and polypropylene. The moisture curable compositions of the present invention include (a) alkoxysilyl capped polymers, (b) alkylsilyl capped polymers, (c) fillers, (d) moisture curing catalyst, and (e) adhesion promoter.

[0015] Useful alkoxysilyl capped polymers include those compounds of the following formula:



I

wherein R is a hydrocarbon diradical which may include heteroatom and/or silicone-containing groups or linkages; A and A' may be each C_{1-30} linear or branched, substituted or unsubstituted aliphatic groups or aromatic-containing groups, with or without interruption by a carboxy, carbamate, carbonate, ureido, urethane or sulfonate linkage; n may be 0 or 1; R^1 and R^2 are substituted or unsubstituted C_{1-12} alkyl or aryl groups; R^3 is a C_{1-12} alkyl, alkenyl, alkoxy, aminoalkyl or aryl group, or a (meth)acryloxyalkyl group.

[0016] Additionally examples of R backbones include alkyd resins, oil modified alkyd resins, unsaturated polyesters, natural oils, (e.g., linseed, tung, soybean), epoxides, nylons, thermoplastic polyester (e.g., polyethyleneterephthalate, polybutyleneterephthalate), polycarbonates, i.e., thermoplastic and thermoset, polyethylenes, polybutylenes, polystyrenes, polypropylenes, ethylene propylene co- and terpolymers, acrylics (homopolymer and copolymers of acrylic acid, acrylates, mathacrylates, acrylamides, their salts, hydrohalides, etc.), phenolic resins, polyoxymethylene (homopolymers and copolymers), polyethylene oxides, polypropylene oxides, polybutylene oxides or polytetramethylene oxides, polyurethanes, polysulfones, polysulfide rubbers, nitrocelluloses, vinyl butyrates, vinyls (vinyl chloride and/or vinyl acetate containing polymers), ethyl cellulose, the cellulose acetates and butyrates, viscose rayon, shellac,

waxes, ethylene copolymers (e.g., ethylenevinyl acetate copolymers, ethylene-acrylic acid copolymers, ethyleneacrylate copolymers), organic rubbers, silicone greases, resins and rubbers and the like.

[0017] R may also include natural rubber; those formed from the homopolymerization of butadiene and its homologues and derivatives such as: cis-1,4-polyisoprene; 3,4-polyisoprene; cis-1,4-polybutadiene; trans-1,4-polybutadiene; 1,2-polybutadiene; and those formed from the copolymerization of butadiene and its homologues and derivatives with one or more copolymerizable monomers containing ethylenic unsaturation such as styrene and its derivatives, vinyl-pyridine and its derivatives, acrylonitrile, isobutylene and alkyl-substituted acrylates such as methylmethacrylate. Examples include styrene-butadiene copolymer rubber composed of various percentages of styrene and butadiene and employing the various isomers of butadiene as desired (hereinafter "SBR"); terpolymers of styrene, isoprene and butadiene polymers, and their various isomers; acrylonitrile-based copolymer and terpolymer rubber compositions; and isobutylene-based rubber compositions; or a mixture thereof, as described in, for example, U.S. Pat. Nos. 4,530,959; 4,616,065; 4,748,199; 4,866,131; 4,894,420; 4,925,894; 5,082,901; and 5,162,409.

[0018] Other suitable organic polymers useful as R backbones are copolymers of ethylene with other high alpha olefins such as propylene, butene-1 and pentene-1 and a diene monomer. The organic polymers may be block, random, or sequential and may be prepared by emulsion (e.g. e-SBR) or solution polymerization processes (e.g. s-SBR). Additional polymers which may be used include those which are partially or fully functionalized including coupled or star-branched polymers. Additional specific examples of functionalized organic rubbers include polychloroprene, chlorobutyl and bromobutyl rubber as well as brominated isobutylene-co-paramethylstyrene rubber. The preferred organic rubbers are polybutadiene, s-SBR and mixtures thereof.

[0019] Silicone rubbers which are useful as R include organic polysiloxane compositions in which the organic polysiloxane is linear or branched, and optionally may contain, in addition to the hydrocarbon groups, certain reactive groups such as for example, hydroxyl, hydrolyzable

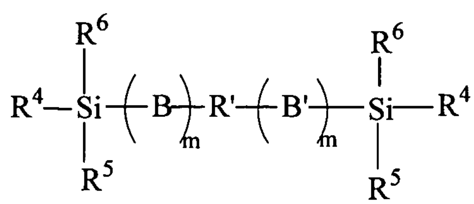
groups, alkenyl groups such as vinyl, hydrogen, fluoro, and phenyl. Further examples are given in U.S. Pat. No. 5,009,874, the disclosures of which is, in its entirety, incorporated herein by reference.

[0020] Other useful R backbone polymers include acrylonitrile-butadiene rubber (NBR), fluorine-containing rubber, epychlorohydrin rubber, butyl rubber, halogenated butyl rubber, brominated isobutylene/p-methylstyrene copolymer rubber, chloroprene rubber, ethylene/acrylate copolymer rubber and epoxidized natural rubber, ethylene/propylene/diene rubber (EPDM) and the like. Additional details of useful alkoxysilyl capped polymers are described in WO 02/068501 A2, the contents of which are incorporated herein by reference.

[0021] The alkoxysilyl capped polymers of the present invention desirably have a viscosity in the range of about 10 cps to about 1,000,000 cps and more desirably about 1,000 cps to about 100,000 cps.

[0022] Desirably, in formula I, R represent a polyether such as a polyethylene oxide, polypropylene oxide, polybutylene oxide or polytetramethylene oxide. More desirably, R represents a polypropylene oxide. Polypropylene oxides with a molecular weight from about 6,000 to about 25,000 are useful. Desirably, the polypropylene oxides have a molecular weight from about 10,000 to about 20,000, including molecular weights of about 12,000 and about 18,000. Moreover, trialkoxysilyl capped polymers, where R^3 is a C_{1-12} alkoxy in formula I, are also useful in the present invention.

[0023] Useful alkylsilyl capped plasticizers include those compounds of the following formula:



II

wherein R' is a hydrocarbon diradical which may include heteroatom and/or silicone-containing

groups or linkages; B and B' may be each C_{1-30} linear or branched, substituted or unsubstituted aliphatic groups or aromatic-containing groups, with or without interruption by a carboxy, carbamate, carbonate, ureido, urethane or sulfonate linkage; m may be 0 or 1; R^4 and R^5 are substituted or unsubstituted C_{1-12} alkyl or aryl groups; R^6 is a C_{1-12} alkyl, alkenyl, alkoxy, aminoalkyl or aryl group, or a (meth)acryloxyalkyl group. Moreover, the above-described R backbones may suitably be used for R' in formula II above.

[0024] The alkylsilyl capped polymers of the present invention desirably have a viscosity in the range of about 10 cps to about 1,000,000 cps and more desirably about 1,000 cps to about 100,000 cps.

[0025] Desirably, in formula II, R' is a polyether that represents a polyethylene oxide, polypropylene oxide, polybutylene oxide or polytetramethylene oxide. More desirably, R' represents a polybutylene oxide or polytetramethylene oxide. Polytetramethylene oxides with a molecular weight from about 650 to about 3,000 are useful. Desirably, the polytetramethylene oxides have a molecular weight from about 1,500 to about 2,500. A molecular weight of about 2,000 is also useful. Moreover, trialkylsilyl capped polymers, where R^6 is a C_{1-12} alkyl in formula II, are also useful with the practice of the present invention.

[0026] The present composition may also include a filler component. For example, any commercially available precipitated or ground calcium carbonate can be used with the present invention. The precipitated calcium carbonate should be present, for example, in an amount from about 10 to about 70% by weight of the total composition. Desirably, the calcium carbonate is present in an amount from about 40 to about 70% by weight of the total composition. More desirably, the calcium carbonate is present in an amount from about 50% to about 60% by weight of the total composition.

[0027] Other fillers may also be used with the present invention. These fillers include, for example, lithopone, zirconium silicate, hydroxides, such as hydroxides of calcium, aluminum, magnesium, iron and the like, diatomaceous earth, carbonates, such as sodium,

potassium, and magnesium carbonates, oxides, such as zinc magnesium, chromic, zirconium and aluminum oxides, calcium clay, graphite, synthetic fibers and mixtures thereof.

[0028] The compositions of the present invention are not limited to the above-described alkylsilyl capped plasticizers and other non-alkylsilyl plasticizers, such as aliphatic liquid polymers and oils or silicone plasticizers may be used, when it is desirable for the composition or cured elastomer thereof to have physical properties and characteristics that are modified by inclusion of such a material. Other organic plasticizers that can be used in the present invention include, for example, petroleum derived organic oils. Moreover, other suitable organic plasticizers include, for example, alkyl phosphates, polyalkylene glycol, poly(propylene oxides), hydroxyethylated alkyl phenol, dialkyldithiophosphonate, poly(isobutylenes), poly(olefins) and mixtures thereof. One example of a suitable silicone plasticizer is a triorganosilyl end blocked diorganosiloxane, although other silicone plasticizers can be used provided they do not compromise the adhesive properties of the present composition. Desirably, the plasticizer is a polyethylene powder.

[0029] In the inventive compositions, effective amounts of plasticizers may be added to aid the workability of the final cured elastomer. From about 5 to about 30% by weight of the total composition of plasticizer may be incorporated into the compositions of the present invention. Desirably, from about 5% to about 25% by weight of the total composition of the selected organic plasticizers are incorporated into the compositions of the present invention.

[0030] The compositions of the present invention may also include a moisture cure catalyst, for example dialkyltin dicarboxylates, alkyl titanates and metal alkoxides, to increase the rate of curing, including increasing the rate of condensation crosslinking reactions. Nonlimited examples of moisture cure catalysts include, for example, dibutyltin dilaurate, dibutyltin diacetate, dibutyltin dioctoate, dibutyltin maleate, dialkyl tin hexoate, dioctyltin dilaurate, iron octanoate, zinc octanoate, lead octanoate, cobalt naphthenate, tetrapropyltitanate and tetrabutyltitanate. Other conventional catalysts can also be incorporated into the present invention. For example, the inventive compositions also include an addition cure catalyst. Suitable addition cure catalysts that can be used with the present compositions include platinum-

based ones, such as platinum-siloxane complexes, which facilitate hydrosilation reactions. Elevated temperatures may also be used to cure the inventive compositions of the present invention. The catalysts may be used in an amount within the range of about 0.001% to about 1% by weight of the total composition. Desirably, the catalyst is present from about 0.01% to about 0.1% weight of the total composition. Photoinitiators, such as visible and UV initiators may also be incorporated.

[0031] Desirably, the inventive compositions include an adhesion promoter. Such an adhesion promoter can include, for example, octyl trimethoxysilane, glycidyl trimethoxysilane, methacryloxypropyl trimethoxysilane, vinyl trimethoxysilane, glycidoxypropyltrimethoxysilane, aminopropyltrimethoxysilane, methacryloxypropyltrimethoxy-silane, triallyl-S-tria-zine-2,3,6(1H.3H.5H)-trione aminoethylaminopropyltrimethoxysilane. Desirably, the adhesion promoter is 3-aminopropyltrimethoxysilane. The adhesion promoters, when present, may be used in an amount within the range of about 0.1% to about 10% by weight of the total composition. Desirably, the adhesion promoter is present from about 1% to about 5% by weight on the total composition.

[0032] The inventive compositions cure at room temperature, i.e., from about 30°C to about 40°C, through moisture curing reactions. Increasing the curing temperature of the compositions of the present invention also improve their adhesion to polyolefin substrates, particularly polyethylene and polypropylene substrates. For example, curing temperatures up to about 150°C or greater are useful. Desirably, curing temperatures from about 70°C to about 100°C are used. More desirably, curing temperatures from about 80 to about 85°C, for example 82°C, are useful with the present invention. The above use of elevated temperature should be balanced, of course, with the ability of the underlying substrate to withstand the temperature.

[0033] The compositions in the present invention can include various other components useful in the manufacturing of moisture curable products. For example, various moisture catalysts, fillers, stabilizers, inhibitors, reactive diluents, viscosity modifiers and the like may be incorporated at useful ranges for their intended purposes.

[0034] As depicted in FIGS. 1 and 2, the compositions of the present invention are useful for producing articles of manufacture, especially articles of manufacture that contain polyolefin, for example polyethylene or polypropylene, substrates. An article of manufacture **10** is depicted in FIG. 1. The article **10** includes polyolefin substrate **12** securably attached to a polyolefin substrate **14**. As depicted in FIG. 2, which is a cross section view of the article **10** taken along the 2-2 axis, polyolefin substrates **12** and **14** are securably attached to one and the other by cured adhesive layer **16**. Cured adhesive layer **16** represents the cured compositions of the present invention.

[0035] A method of bonding polyolefin substrates is also included within the scope of the present invention. Polyolefin substrates, for instance polyethylene or polypropylene substrates, are secured to one and the other by applying the inventive compositions therebetween. The inventive compositions adhesively join the substrates upon curing, for example through moisture curing. Adhesion of the inventive compositions to the polyolefin substrates can be further improved by increasing the cure temperature above room temperature, as described above.

[0036] The following non-limiting examples are intended to further illustrate the present invention.

EXAMPLES

Example 1:

[0037] Inventive Compositions were prepared as described in Table 1 below. The triethyloxysilyl capped polyethylene oxide polymer A was prepared by heating a mixture of about 95 weight parts ACCLAIM 12200 (a polypropylene oxide polymer with OH endgroups having a molecular weight of about 12000 from Lyondell) with about 5 weight parts SILQUEST A1310 (isocyanatopropyltriethoxysilane from Crompton) in the presence of dimethyltin dicarboxylate, a tin catalyst, at 35°C to 60°C for about 3 hours. The remaining ingredients were admixed to form Inventive Composition Nos. 1-6, as shown below.

Table 1

Inventive Composition Nos.

Component (wt%):	**1**	**2**	**3**	**4**	**5**	**6**
triethyloxysilyl capped polyethylene oxide	98.7	97.6	61.4	60.7	61.3	60.6
plasticizer			18.4	18.2	18.4	18.2
calcium carbonate filler			18.4	18.2	18.4	18.2
Tin catalyst	0.2	0.2	0.1	0.1	0.2	0.2
vinyl trimethoxy silane		0.6				
vinyl triethoxy silane						
3-aminopropyltrimethoxysilane	1.1	1.6	1.7	2.8	1.5	2.8
3-aminopropyltriethoxysilane						
TOTAL (wt%)	100%	100%	100%	100%	100%	100%

[0038] Inventive Composition Nos. 1-6 were applied to low density polyethylene lap shears (1 ½ inches by 1 inch with a thickness of 0.04 inches). Curing conditions, i.e., cure temperatures and time, were varied and are shown below in Table A-1. All composition cured to the lap shears. Increasing the calcium carbonate filler improved the lap shear strength.

Table A-1
Low-Density Polyethylene (TS210) Lap Shears:
Lap Shear Testing Results

Inventive Composition Nos.	Cure Temp.	Cure Time, days	Shear Strength, psi	Joint Movement, inches	Failure Mode
1	RT	7	2	0.028	AF
2	RT	7	12	0.039	AF
3	RT	7	35	0.032	AF
3	RT/hot	7	37	0.035	AF
4	RT	7	34	0.057	AF
4	RT/hot	7	47	0.045	AF
5	RT	7	30	0.027	AF
5	RT/hot	7	38	0.039	AF
6	RT	7	48	0.047	AF
6	RT/hot	7	36	0.032	AF
1	50°C	7	3	0.029	AF
2	50°C	7	17	0.039	AF
3	50°C	7	38	0.037	AF
4	50°C	7	81	0.093	AF
5	50°C	7	83	0.104	AF
6	50°C	7	93	0.121	AF
3	82°C	7	7	0.013	AF
4	82°C	7	24	0.024	MMF
5	82°C	7	57	0.066	AF
6	82°C	7	92	0.110	MMF

Notes:
RT: room temperature
AF: adhesive failure
CF: cohesive failure
MMF: mixed mode of failure (partial AF and partial CF)

[0039] Inventive Composition Nos. 1-6 were applied to high density polyethylene lap shears (1 ½ inches by 1 inch with a thickness of 0.04 inches). Curing conditions, i.e., cure temperatures and time, were varied and are shown below in Table B-1. All composition cured to the lap shears. Increasing the calcium carbonate filler generally improved the lap shear strengths, particularly at room temperature curing conditions.

Table B-1
High-Density Polyethylene (TS227) Block Shears:
Lap Shear Testing Results

Inventive Composition Nos.	Cure Temp.	Cure Time, days	Shear Strength, psi	Joint Movement, inches	Failure Mode
1	RT	7	74	0.042	AF
2	RT	7	85	0.030	AF
3	RT	7	68	0.034	AF
3	RT/hot	7	78	0.040	AF
4	RT	7	70	0.021	AF
4	RT/hot	7	85	0.026	AF
5	RT	7	90	0.040	AF
5	RT/hot	7	82	0.023	AF
6	RT	7	88	0.043	MMF
6	RT/hot	7	84	0.024	AF
1	50°C	7	140	0.061	MMF
2	50°C	7	203	0.076	MMF
3	50°C	7	79	0.040	AF
4	50°C	7	94	0.041	MMF
5	50°C	7	85	0.035	AF
6	50°C	7	128	0.059	MMF
3	82°C	7	13	0.010	MMF
4	82°C	7	52	0.011	MMF
5	82°C	7	70	0.022	MMF
6	82°C	7	7	0.008	MMF

Notes:
RT: room temperature
AF: adhesive failure
CF: cohesive failure
MMF: mixed mode of failure (partial AF and partial CF)

[0040] Inventive Composition Nos. 1-6 were applied to polypropylene lap shears (1 ½ inches by 1 inch with a thickness of 0.04 inches). Curing conditions, i.e., cure temperatures and time, were varied and are shown below in Table C-1. All composition cured to the lap shears. Increasing the calcium carbonate filler generally improved the lap shear strengths, particularly at room temperature curing conditions.

Table C-1
Polypropylene (TS226) Block Shears:
Lap Shear Testing Results

Inventive Composition Nos.	Cure Temp.	Cure Time, days	Shear Strength, psi	Joint Movement, inches	Failure Mode
1	RT	7	17	0.018	AF
2	RT	7	48	0.019	AF
3	RT	7	37	0.012	AF
3	RT/hot	7	40	0.012	AF
4	RT	7	20	0.010	AF
4	RT/hot	7	39	0.008	AF
5	RT	7	42	0.011	AF
5	RT/hot	7	17	0.006	AF
6	RT	7	40	0.012	AF
6	RT/hot	7	61	0.032	AF
1	50°C	7	29	0.016	AF
2	50°C	7	49	0.019	AF
3	50°C	7	26	0.009	AF
4	50°C	7	65	0.016	AF
5	50°C	7	22	0.008	AF
6	50°C	7	54	0.014	AF
3	82°C	7	10	0.010	M
4	82°C	7	44	0.013	AF
5	82°C	7	7	0.008	AF
6	82°C	7	8	0.009	AF

Notes:
RT: room temperature
AF: adhesive failure
CF: cohesive failure
MMF: mixed mode of failure (partial AF and partial CF)

Example 2:

[0041] Inventive Compositions were prepared as described in Table 2 below. The triethyloxysilyl capped polyethylene oxide polymer was prepared by heating a mixture of about 95 weight parts ACCLAIM 12000 (a polypropylene oxide polymer with OH endgroups having a molecular weight of about 12000 from Lyondell) with about 5 weight parts SILQUEST A1310 (isocyanatopropyltriethoxysilane from Crompton) in the presence of dimethyltin dicarboxylate, a tin catalyst, at 35°C to 60°C for about 3 hours. The trimethylsilyl capped polytetramethylene oxide polymer was prepared by capping polytetramethylene ether glycol (POLYMEG 2000 from Lyondell) with tetramethylxylylene diisocyanate (TMXDI) and n-ethylaminopropyltrimethoxy-silane. The remaining ingredients were admixed to form Inventive Compositions Nos. 7-13, as shown below.

Table 2

Inventive Composition Nos.

Component (wt%):	7	8	9	10	11	12	13
triethyloxysilyl capped polyethylene oxide	14.3	14.1					9.3
trimethylsilyl capped polytetramethylene oxide			17.5	17.2	20.4	20.1	10.0
plasticizer	14.3	14.1	17.5	17.2	15.9	15.8	17.1
calcium carbonate filler	47.6	46.9	63.0	62.1	44.3	43.8	52.8
polyethylene powder	22.1	22.1			17.7	17.5	8.7
Tin catalyst	0.1	0.1	0.1	0.1	0.1	0.1	0.1
vinyl trimethoxy silane			0.4	0.4			
vinyl triethoxy silane							
3-aminopropyl-trimethoxysilane	1.6	2.7	1.5	3.0	1.6	2.7	2.0
3-aminopropyl-triethoxysilane							
TOTAL (wt%)	100%	100%	100%	100%	100%	100%	100%

[0042] Inventive Composition Nos. 7-13 were applied to low density polyethylene lap shears (1 ½ inches by 1 inch with a thickness of 0.04 inches). Curing conditions, i.e., cure temperatures and time, were varied and are shown below in Table A-2. All composition cured to the lap shears. The incorporation of calcium carbonate filler and an adhesion promoter, i.e., aminopropyltrimethoxysilane, improved the lap shear strength. Moreover, the inclusion of both the triethyloxysilyl capped polyethylene oxide polymer and the trimethylsilyl capped polytetramethylene oxide plasticizer improved the lap shear strength.

Table A-2
Low-Density Polyethylene (TS210) Lap Shears:
Lap Shear Testing Results

Inventive Composition Nos.	Cure Temp.	Cure Time, days	Shear Strength, psi	Joint Movement, inches	Failure Mode
7	RT	8	67	0.071	MMF
8	RT	8	72	0.078	MMF
9	RT	7	70	0.082	AF
9	RT	17	101	0.123	AF
10	RT	7	53	0.058	AF
10	RT	17	96	0.101	AF
11	RT	10	38	0.030	AF
11	RT	14	70	0.073	AF
12	RT	10	70	0.072	AF
12	RT	14	100	0.102	AF
13	RT	7	48	0.050	MMF
7	50°C/RT	7/7	82	0.112	AF
8	50°C/RT	7/7	68	0.095	AF
11	50°C	10	68	0.080	AF
12	50°C	10	87	0.094	AF
7	82°C/RT	4/4	148	0.173	CF
8	82°C/RT	4/4	139	0.233	CF
9	82°C/RT	3/4	118	0.203	MMF
9	82°C	7	108	0.219	AF
9	82°C	17	144	0.206	AF
10	82°C	7	119	0.224	AF
10	82°C	17	69	0.080	AF
11	82°C	11	135	0.261	MMF
11	82°C°	25	125	0.141	AF
12	82°C	11	95	0.105	AF
12	82°C	25	100	0.115	AF
13	82°C/RT	7	112	0.195	MMF

Notes:
RT: room temperature
AF: adhesive failure
CF: cohesive failure
MMF: mixed mode of failure (partial AF and partial CF)

[0043] Inventive Composition Nos. 7-13 were applied to high density polyethylene lap shears (1 ½ inches by 1 inch with a thickness of 0.04 inches). Curing conditions, i.e., cure temperatures and time, were varied and are shown below in Table B-2. All composition cured to the lap shears. The incorporation of calcium carbonate filler and an adhesion promoter, i.e., aminopropyltrimethoxysilane, improved the lap shear strength. Moreover, the inclusion of both the triethyloxysilyl capped polyethylene oxide polymer and the trimethylsilyl capped polytetramethylene oxide plasticizer improved the lap shear strength.

Table B-2
High-Density Polyethylene (TS227) Block Shears:
Lap Shear Testing Results

Inventive Composition Nos.	Cure Temp.	Cure Time, days	Shear Strength, psi	Joint Movement, inches	Failure Mode
7	RT	8	109	0.027	MMF
8	RT	8	56	0.013	AF
9	RT	7	47	0.021	AF
9	RT	17	57	0.014	MMF
10	RT	7	74	0.019	AF
10	RT	17	171	0.028	AF
11	RT	10	73	0.021	AF
11	RT	14	76	0.022	AF
12	RT	10	96	0.018	AF
12	RT	14	184	0.030	AF
13	RT	7	65	0.026	MMF
7	50°C/RT	7/7	83	0.023	MMF
8	50°C/RT	7/7	91	0.025	MMF
11	50°C	10	134	0.027	AF
12	50°C	10	201	0.031	AF
7	82°C/RT	4/4	63	0.018	80% CF
8	82°C/RT	4/4	32	0.009	AF
9	82°C/RT	3/4	176	0.049	MMF
9	82°C	7	183	0.031	AF
9	82°C	17	170	0.036	AF
10	82°C	7	213	0.025	MMF
10	82°C	17	212	0.024	MMF
11	82°C	11	150	0.032	MMF
11	82°C	25	63	0.023	AF
12	82°C	11	179	0.025	MMF
12	82°C	25	129	0.024	AF
13	82°C/RT	7	240	0.041	MMF

Notes:
RT: room temperature
AF: adhesive failure
CF: cohesive failure
MMF: mixed mode of failure (partial AF and partial CF)

[0044] Inventive Composition Nos. 7-13 were applied to polypropylene lap shears (1 ½ inches by 1 inch with a thickness of 0.04 inches). Curing conditions, i.e., cure temperatures and time, were varied and are shown below in Table C-2. All composition cured to the lap shears. The incorporation of calcium carbonate filler and an adhesion promoter, i.e., aminopropyl-trimethoxysilane, improved the lap shear strength. Moreover, the inclusion of both the triethyloxysilyl capped polyethylene oxide polymer and the trimethylsilyl capped polytetramethylene oxide plasticizer improved the lap shear strength.

Table C-2
Polypropylene (TS226) Block Shears:
Lap Shear Testing Results

Inventive Composition Nos.	Cure Temp.	Cure Time, days	Shear Strength, psi	Joint Movement, inches	Failure Mode
7	RT	8	53	0.009	AF
8	RT	8	86	0.016	MMF
9	RT	7	28	0.008	AF/90%CF
9	RT	17			
10	RT	7	36	0.009	AF/55%CF
10	RT	17			
11	RT	10	46	0.012	AF
11	RT	14	58	0.015	AF
12	RT	10	86	0.018	AF
12	RT	14	11	0.014	AF
13	RT	7	15	0.008	AF
7	50°C/RT	7/7	83	0.019	AF
8	50°C/RT	7/7	86	0.017	AF
11	50°C	10	149	0.025	AF
12	50°C	10	31	0.0006	AF
7	82°C/RT	4/4	84	0.014	MMF
8	82°C/RT	4/4	95	0.018	AF
9	82°C/RT	3/4	72	0.016	AF
9	82°C	7	35	0.012	AF
9	82°C	17	50	0.022	AF
10	82°C	7	29	0.007	AF
10	82°C	17	63	0.012	AF
11	82°C	11	115	0.021	AF
11	82°C	25	144	0.024	AF
12	82°C	11	84	0.012	AF
12	82°C	25	61	0.014	AF
13	82°C/RT	7	205	0.026	MMF

Notes:
RT: room temperature
AF: adhesive failure
CF: cohesive failure
MMF: mixed mode of failure (partial AF and partial CF)

Example 3:

[0045] Inventive Compositions were prepared as described in Table 3 below. The triethyloxysilyl capped polyethylene oxide polymer was prepared by heating a mixture of about 95 weight parts ACCLAIM 18000 (a polypropylene oxide polymer with OH endgroups having a molecular weight of about 18000 from Lyondell) with about 5 weight parts SILQUEST A1310 (isocyanatopropyltriethoxysilane from Crompton) in the presence of dimethyltin dicarboxylate, a tin catalyst, at 35°C to 60°C for about 3 hours. The trimethylsilyl capped polytetramethylene oxide polymer was prepared by capping polytetramethylene ether glycol (POLYMEG 2000 from Lyondell) with tetramethylxylylene diisocyanate (TMXDI) and n-ethylaminopropyltrimethoxy-silane. The remaining ingredients were admixed to form Inventive Compositions Nos. 14-18, as shown below.

Table 3

Inventive Composition Nos.

Component (wt%):	**14**	**15**	**16**	**17**	**18**
triethyloxysilyl capped polyethylene oxide	49.4	24.7			10.0
trimethylsilyl capped polytetramethylene oxide	10.1	34.8	59.5	28.0	28.0
calcium carbonate filler	21.9	21.9	21.9		
polyethylene powder	8.7	8.7	8.7	50.0	50.0
Plasticizer	7.9	7.9	7.9	20.0	10.0
3-aminopropyltrimethoxysilane	1.9	1.9	1.9	1.9	1.9
tin catalyst	0.1	0.1	0.1	0.1	0.1
TOTAL (wt%)	100%	100%	100%	100%	100%

[0046] Inventive Composition Nos. 14-18 were applied to low density polyethylene lap shears (1 ½ inches by 1 inch with a thickness of 0.04 inches). Curing conditions, i.e., cure temperatures and time, were varied and are shown below in Table A-3. All composition cured to the lap shears. Increasing the calcium carbonate filler improved the lap shear strength. Moreover, the inclusion of both the triethyloxysilyl capped polyethylene oxide polymer and the trimethylsilyl capped polytetramethylene oxide plasticizer improved the lap shear strength.

Table A-3
Low-Density Polyethylene (TS210) Lap Shears:
Lap Shear Testing Results

Inventive Composition Nos.	Cure Temp.	Cure Time, days	Shear Strength, psi	Joint Movement, inches	Failure Mode
14	RT	7	23	0.035	AF
15	RT	16			
16	RT	16	46	0.031	AF
14	82°C/RT	7	41	0.073	AF
15	82°C	6	47	0.049	AF
15	82°C	13	46	0.044	AF
16	82°C	6	26	0.037	AF
16	82°C	13	46	0.049	AF
17	82°C	7	35	0.039	CF
17	82°C	14	44	0.034	CF
18	82°C	7	66	0.079	MMF
18	82°C	14	82	0.077	CF

Notes:
RT: room temperature
AF: adhesive failure
CF: cohesive failure
MMF: mixed mode of failure (partial AF and partial CF)

[0047] Inventive Composition Nos. 14-18 were applied to high density polyethylene lap shears (1 ½ inches by 1 inch with a thickness of 0.04 inches). Curing conditions, i.e., cure temperatures and time, were varied and are shown below in Table B-3. All composition cured to

the lap shears. Increasing the calcium carbonate filler generally improved the lap shear strengths, particularly at room temperature curing conditions. Moreover, the inclusion of both the triethyloxysilyl capped polyethylene oxide polymer and the trimethylsilyl capped polytetramethylene oxide plasticizer improved the lap shear strength.

Table B-3
High-Density Polyethylene (TS227) Block Shears:
Lap Shear Testing Results

Inventive Composition Nos.	Cure Temp.	Cure Time, days	Shear Strength, psi	Joint Movement, inches	Failure Mode
14	RT	7	66	0.019	AF
15	RT	16	86	0.017	AF
16	RT	16	132	0.021	AF
14	82°C/RT	7	89	0.035	AF
15	82°C	6	70	0.018	AF
15	82°C	13	107	0.041	AF
16	82°C	6	101	0.025	AF
16	82°C	13	103	0.023	AF
17	82°C	7	29	0.014	CF
17	82°C	14	22	0.022	CF
18	82°C	7	48	0.014	CF
18	82°C	14	67	0.025	CF

Notes:
RT: room temperature
AF: adhesive failure
CF: cohesive failure
MMF: mixed mode of failure (partial AF and partial CF)

[0048] Inventive Composition Nos. 14-18 were applied to polypropylene lap shears (1 ½ inches by 1 inch with a thickness of 0.04 inches). Curing conditions, i.e., cure temperatures and time, were varied and are shown below in Table C-3. All composition cured to the lap shears. Increasing the calcium carbonate filler generally improved the lap shear strengths, particularly at room temperature curing conditions. Moreover, the inclusion of both the triethyloxysilyl capped

polyethylene oxide polymer and the trimethylsilyl capped polytetramethylene oxide plasticizer improved the lap shear strength.

Table C-3
Polypropylene (TS226) Block Shears:
Lap Shear Testing Results

Inventive Composition Nos.	Cure Temp.	Cure Time, days	Shear Strength, psi	Joint Movement, inches	Failure Mode
14	RT	7	8	0.043	AF
15	RT	16	30	0.008	AF
16	RT	16	72	0.012	AF
14	82°C/RT	7	33	0.014	AF
15	82°C	6	64	0.023	AF
15	82°C	13	21	0.015	AF
16	82°C	6	2	0.013	AF
16	82°C	13	54	0.015	AF
17	82°C	7	28	0.018	CF
17	82°C	14	22	0.019	CF
18	82°C	7	40	0.014	AF
18	82°C	14	63	0.024	CF

Notes:
RT: room temperature
AF: adhesive failure
CF: cohesive failure
MMF: mixed mode of failure (partial AF and partial CF)

Example 4:

[0049] Inventive Compositions were prepared as described in Table 4 below. A polydimethylsiloxane with vinyldimethoxysilyl groups having a viscosity of about 5000 mPas and the remaining ingredients were admixed to form Inventive Compositions Nos. 19-22, as shown below.

Table 4

Inventive Composition Nos.

Component (wt%):	19	20	21	22
polydimethylsiloxane with vinyldimethoxysilyl groups	40.5	33.5	30.6	32.8
calcium carbonate filler		44.6	56.8	65.5
polyethylene powder	57.7	20.1	10.9	
3-aminopropyltrimethoxysilane	1.7	1.7	1.6	1.6
tin catalyst	0.1	0.1	0.1	0.1
TOTAL (wt%)	100%	100%	100%	100%

[0050] Inventive Composition Nos. 19-22 were applied to low density polyethylene lap shears (1 ½ inches by 1 inch with a thickness of 0.04 inches). Curing conditions, i.e., cure temperatures and time, were varied and are shown below in Table A-4. All composition cured to the lap shears. Increasing the calcium carbonate filler improved the lap shear strength.

Table A-4
Low-Density Polyethylene (TS210) Lap Shears:
Lap Shear Testing Results

Inventive Composition Nos.	Cure Temp.	Cure Time, days	Shear Strength, psi	Joint Movement, inches	Failure Mode
19	82°C	6	46	0.052	CF
19	82°C	13	44	0.035	CF
20	82°C	8	157	0.256	90% CF
21	82°C	5	98	0.086	AF
22	82°C	5	170	1.000	CF

Notes:
AF: adhesive failure
CF: cohesive failure
MMF: mixed mode of failure (partial AF and partial CF)

[0051] Inventive Composition Nos. 19-22 were applied to high density polyethylene lap shears (1 ½ inches by 1 inch with a thickness of 0.04 inches). Curing conditions, i.e., cure temperatures and time, were varied and are shown below in Table B-4. All composition cured to the lap shears. Increasing the calcium carbonate filler generally improved the lap shear strengths, particularly at room temperature curing conditions.

Table B-4
High-Density Polyethylene (TS227) Block Shears:
Lap Shear Testing Results

Inventive Composition Nos.	Cure Temp.	Cure Time, days	Shear Strength, psi	Joint Movement, inches	Failure Mode
19	82°C	6	31	0.008	CF
19	82°C	13	45	0.015	CF
20	82°C	8	165	0.049	CF
21	82°C	5	217	0.040	30%CF
22	82°C	5	279	0.053	AF

Notes:
AF: adhesive failure
CF: cohesive failure
MMF: mixed mode of failure (partial AF and partial CF)

[0052] Inventive Composition Nos. 19-22 were applied to polypropylene lap shears (1 ½ inches by 1 inch with a thickness of 0.04 inches). Curing conditions, i.e., cure temperatures and time, were varied and are shown below in Table C-4. All composition cured to the lap shears. Increasing the calcium carbonate filler generally improved the lap shear strengths, particularly at room temperature curing conditions.

Table C-4
Polypropylene (TS226) Block Shears:
Lap Shear Testing Results

Inventive Composition Nos.	Cure Temp.	Cure Time, days	Shear Strength, psi	Joint Movement, inches	Failure Mode
19	82°C	6	32	0.013	CF
19	82°C	13	39	0.018	CF
20	82°C	8	46	0.017	AF
21	82°C	5	137	0.031	AF
22	82°C	5	20	0.011	AF

Notes:
AF: adhesive failure
CF: cohesive failure
MMF: mixed mode of failure (partial AF and partial CF)

[0053] While the invention has been described in reference to various aspects and embodiments, it will be appreciated that the invention is not limited by these, but may be subject to numerous variations, modifications and other embodiments, all of which are contemplated within the spirit and scope of the invention as claimed.